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# Measurement and analysis of isobaric contributions in pre-equilibrium nuclear reactions

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**Abstract** : Excitation functions of the reactions $^{169}$Tm [(α,p3n), (α,αn), (α,α2n) and (α,α4n)] are investigated upto 60 MeV using foil stack activation technique and Ge(Li) gamma ray spectroscopy method. Isobaric contributions from the decaying parent and grand parent nuclei to the final product nucleus of interest, is discussed and separated out wherever necessary, for which a mathematical formulation from first principle is developed. The measured cross sections are compared with the updated version of hybrid model using initial exciton configuration $n_0 = 4$ (4*p*0*h*). While the model predictions give an approximate agreement at higher energies in (α,pxn) reactions, they are equally bad for all (α, αxn) type of reactions for which some direct reaction contributions are indicated.

**Keywords** : Pre-equilibrium nuclear reactions, isobaric contributions, excitation function

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## 1. Introduction

In recent years, a great deal of efforts have been made for the development of a variety of pre-equilibrium models in order to explain high energy continuum of the ejectiles in the intermediate energy-nuclear reactions. Existing pre-equilibrium models are originally designed to describe the emission of nucleons. They calculate particle emission rates by applying reciprocity to the ejectiles under consideration, rather than the system as a whole. This does not pose much of a problem as long as only nucleons are considered, since their existence inside the nuclei are generally accepted and since there is reasonable theoretical guidance to follow their behaviour (single particle state densities, average potential, Pauli restrictions *etc*). When it comes to clusters, however, no information about pre-formation and behaviour inside the nucleus is available on an *a priori* basis. Thus pre-equilibrium models to describe cluster emission have introduced parameters or assumptions to fill the gap. Most notably, the pre-formation parameter and the assumptions employed for a cluster

of single particle state density have provided powerful tools to reproduce experimental data. Several such approaches [1–3] have been suggested using this concept. Cline [4] suggested a crude model of statistical coalescence which was subsequently improved and corrected by the group at Bratislava [5]. Machner [6] followed this approach and interpreted its intrinsic cluster pre-formation probabilities in terms of the coalescence model originally described for high energy heavy ion reactions.

There are a few complications encountered in the experimental study of ($\alpha$, $x$n$z\alpha$) type of reactions using activation technique, such as isobaric precursor contribution to the actual residual nucleus whose activity is being studied to determine the cross section. Such situation is often encountered in the activation method for the determination of cross sections of [($\alpha$, ($x$+4)n)], [$\alpha$, p($x$+3)n] and ($\alpha$, $\alpha$n) reactions leading to the residual nuclei which are isobars of one another and form a $\beta$ decaying chain. Thus, the growth and decay of last residual nucleus is influenced by the contributions coming from its isobaric precursors. In such situation, the simple cross section formula [7–9] cannot be employed to determine the cross section of residual nucleus, since it does not take into account the contributions from the decaying parent and/or grand parent nuclei (these include the contributions during their growth throughout the period of irradiation and later due to their decay). In such cases, the cross sections of the parent nucleus is first determined using the expression given in Section 2.1; and later the cross section of the daughter nucleus is calculated. A mathematical formulation from first principle is developed to separate out exactly point by point, the isobaric precursor contributions from the measured inclusive excitation function which are given in Section 2.2.

In the work reported here, a systematic reinvestigation of $^{169}$Tm ($\alpha$,p3n), $^{169}$Tm ($\alpha$, $x\alpha x$n) ; $x$ = 1, 2, 4 is made. To the best of the knowledge of the author, one of these reactions namely $^{169}$Tm ($\alpha$, p3n) reaction was earlier studied by Sau *et al* [10] employing a NaI detector having 8% resolution, upto a beam energy of 50 MeV. Since in their study, they have not taken into account the isobaric contribution of the decaying parent nucleus, the overwhelming large contribution of $^{169}$Lu formed in $^{169}$Tm($\alpha$,4n) reaction is predominant and they have in fact, measured the $^{169}$Tm($\alpha$,4n) reaction only. Their obvious conclusion was that the $^{169}$Tm($\alpha$,p3n) reaction could not be studied by the activation technique because it would be masked by the combination of $^{169}$Tm($\alpha$,4n) reaction. In this scenario, it is felt worthwhile (i) to carry out these studies with improved accuracy, extending the measurements to higher energies by employing high resolution HPGe detector having 2 KeV resolution for 1332 keV of $^{60}$Co and, (ii) to employ appropriate formula for cross section determination wherever required (iii) and to compare the measured cross section with updated pre-equilibrium hybrid model using $n_0 = 4(4p0h)$ to give a meaningful picture of reaction mechanism.

## 2. Experimental

Excitation function of $^{169}$Tm [($\alpha$, p3n), ($\alpha$, $\alpha$n), ($\alpha$, $\alpha$2n) and ($\alpha$, $\alpha$4n)] reactions are measured upto 60 MeV. Self supporting Thulium foils of thicknesses 29 mg/cm$^2$

targets together with several aluminium degraders of varying thickness are used in the stack.

Alpha irradiation has been made with 60 MeV alpha particles at Variable Energy Cyclotron Centre, Calcutta, India. Beam current of the order of 200 nA is maintained in the stack. The experimental technique and procedure adopted in the present work is the same as mentioned in our earlier work [11–14]. Spectroscopic information [15] about various residual nuclei is given in Table 1. The energy and efficiency calibration of the detector are

**Table 1**. Decay characteristics of the nuclides investigated

| Reaction | Residual nucleus | $Q$-value (MeV) | $T_{1/2}$ | $E_\gamma$ (keV) | $I_\gamma$ (keV) |
|---|---|---|---|---|---|
| $^{169}$Tm(α,p3n) | $^{169}$Yb | −30 0 | 32.02d | 110 | 18.0 |
| | | | | 130 | 11.4 |
| | | | | 177 | 21.7 |
| $^{169}$Tm(α,αn) | $^{168}$Tm | −8.0 | 93.10d | 447 | 22.0 |
| | | | | 720 | 11.0 |
| | | | | 741 | 11 3 |
| | | | | 816 | 46.3 |
| $^{169}$Tm(α,α2n) | $^{167}$Tm | −14.87 | 9.25d | 208 | 41.0 |
| | | | | 532 | 1.6 |
| $^{169}$Tm(α,α4n) | $^{165}$Tm | −30.63 | 30.06hr | 243 | 35.0 |
| | | | | 297 | 24 7 |
| | | | | 1397 | 9.3 |

performed with a standard calibrated $^{152}$Eu source obtained from Radio Chemistry Division of VECC. The over all error in the present measurements ranges between 8–12% which includes photo-peak area, detection efficiency, uniformity of foil thickness and that of monitor reaction cross section.

*2.1. Cross section determination :*

The number of observed decays $y(t)$ is related to the total number of decays $z(t)$ during the measuring time $t$ by

$$Z(t) = \frac{y(t)}{[\varepsilon(En)\, In(\text{abs})]},$$

where $\varepsilon(En)$ is the detector efficiency and $In$ (abs) is the absolute γ-ray abundance (yield) per decay. The corresponding reaction yield $N_0$. is given for simple decays (simple decays corresponds to direct production of radio isotopes by the nuclear reactions) by

$$N_o = \frac{Z(t)\, e^{\lambda t_2}}{\left\{\left[1 - e^{-\lambda t_2}\right]\left[1 - e^{-\lambda t_1}\right] / \lambda t_1\right\}},$$

where $\lambda = \ln 2/T_{1/2}$ is the decay constant and $t_1$ and $t_2$ are the time of irradiation and waiting time respectively. $N_0$ is related to the cross section $\sigma$ by the relation,

$$N_o = \sigma (N_{av}\, \delta x)\, I, \tag{1}$$

where $N_{av}$ is the number of atoms/cm$^3$ of the target material, $\delta x$ is the thickness of the foil in cms, $I$ is the flux of the incident particle (Integrated).

*2.2. Formalism for isobaric contribution :*

As mentioned in the introduction, the above expression is valid for straight-forward cases. But more often the nuclear reactions in which two genetically related product nuclei (isobars or isomers) are formed with comparable half lives and one of them, the daughter nucleus, being continuously fed by the mother nucleus by radioactive decay. In such cases, the expression given above cannot be employed to determine the cross section of the daughter nucleus, as it does not take into account the contribution from feeding precursor nucleus. Therefore, the cross section $\sigma_1$ of the mother nucleus is first determined using eq. (1). The cross section $\sigma_2$ of the daughter nucleus is determined using the expression given below :

$$\begin{aligned}\frac{A_\gamma}{P_\gamma \theta_\gamma} = N_T \phi \Bigg[ & \frac{\sigma_1 \lambda_2}{\lambda_2 - \lambda_1} \left(1 - e^{-\lambda_1 t_1}\right) \left\{ \frac{e^{-\lambda_1 tw} - e^{-\lambda_1 (tw+\Delta)}}{\lambda_1} \right. \\ & \left. - \frac{e^{-\lambda_2 tw} - e^{-\lambda_2 (tw+\Delta)}}{\lambda_2} \right\} + \frac{\sigma_1 + \sigma_2}{\lambda_2} \left(1 - e^{-\lambda_2 t_1}\right) \\ & \times \left\{ e^{-\lambda_2 tw} - e^{-\lambda_2 (tw+\Delta)} \right\} + \frac{\sigma_1}{\lambda_2 - \lambda_1} \left\{ \left(e^{-\lambda_1 t_2} - e^{-\lambda_2 t_2}\right) \right. \\ & \left. \times \left\{ e^{-\lambda_2 tw} - e^{-\lambda_2 (tw+\Delta)} \right\} \right] \end{aligned} \tag{2}$$

where suffix 1 stands for mother and 2 stands for daughter nucleus.

## 3. Results and discussion

The study of excitation function is generally complicated by the possibility of isobaric precursors contributing to the cross section of the formation of the residual nucleus of interest. The applicability of the mathematical formulation for the isobaric contribution to the reaction residues of interest and their salient features are discussed below :

*3.1. $^{169}$Tm(α,p3n) $^{169}$Yb reaction :*

In the present investigation, the excitation function for this reaction is exclusively and carefully determined employing the activation technique and the above formalism. At first, the cross sections of this reaction for various projectile energies is calculated using eq. (1) and is found that the $\sigma_{max}$ of this reaction is of the order of 1000 mb. This is because of the large contribution of $^{169}$Lu formed in (α,4n) reactions. This interfering contribution in the measurements of (α,p3n) cross section from that of (α,4n) reaction is really a major

problem, especially in view of the fact that the later cross section is generally 10 times larger than the former, which is in question. The major part of this effect is of course, due to the Coulomb field hindrance to the protons.

It is precisely for this reason, Sau *et al* [10] could not succeed in studying the excitation function of $^{169}$Tm(α, ,p3n) reaction although they made an unsuccessful attempt by measuring the activity of the product nucleus $^{169}$Yb ($T_{1/2}$ = 32 d). Thus, taking the experimental cross section of (α,4n) reaction from our earlier work [12], true cross section of (α,p3n) reaction are calculated using the expression in eq. (2). Figure 1 shows the comparison of the present experimental results for $^{169}$Tm(α,p3n) reaction with the updated

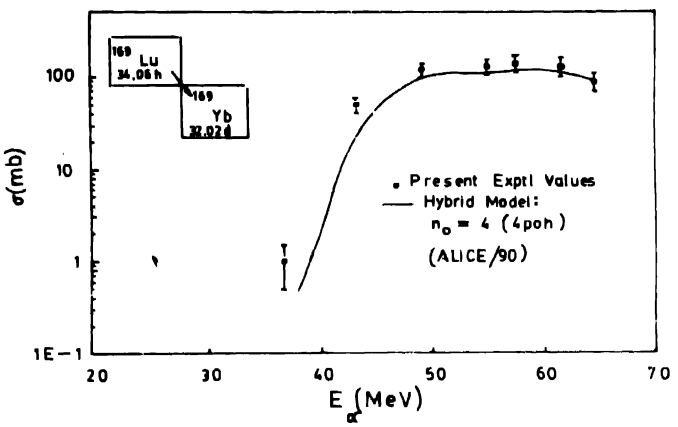


**Figure 1.** Excitation function of $^{169}$Tm(α,p3n)$^{169}$Yb reaction

hybrid model using $n_0$ = 4(4*p*0*h*). It may be observed that in the low energy part, the compound nucleus peak is absent. The broad excitation function may indicate a mixture of equilibrium and pre-equilibrium effects in this energy region. For this reason, it can be observed that hybrid model predictions are insensitive to the initial exciton number. Therefore, except to recognize the influence of pre-equilibrium effects in the energy region 50–60 MeV, no definite conclusion can be drawn.

### *3.2. $^{169}$Tm(α, αn) $^{168}$Tm reaction :*

As already stated, the study of (α,αxn) type of reactions is difficult and interesting at the same time. They have generally low cross section so that their characteristic gamma rays are masked by the compton background due to much stronger gamma rays coming from (α,xn) type of reactions on the same target. The interest in this type of reaction is centered around the mechanism of alpha emission. The basic assumption such as the existence of 'preformed' alpha particles in nucleus and the ideas of 'coalescence' models have to be tested against detailed experimental studies on (α,αxn) type of reaction. However, as pointed out previously, their study by activation technique, as far as the excitation function is concerned, is generally complicated by the possibility of two isobaric precursors one coming from (α,p*x*'n) and the other (α,α*x*"n) reactions. But in specific case such as

$^{169}Tm(\alpha,\alpha n)^{168}Tm$ reaction, such isobaric precursor contributions are absent simply because the immediate precursor, $^{168}Yb$, happens to be a stable nucleus.

A theoretical comparison is shown in Figure 2 for the reaction $Tm(\alpha,\alpha n)$ with the prediction of updated hybrid model. It can be seen that the theoretical predictions are lower by more than an order of magnitude, compared to the experimental ones. In fact the shape of the excitation function is completely different with a very broad peak occurring in the energy region where a deep valley is indicated by the theoretical predictions. This is of course not surprising because hybrid model is not designed to deal with alpha particle emission in the pre-equilibrium phase which is quite likely at moderate energies.

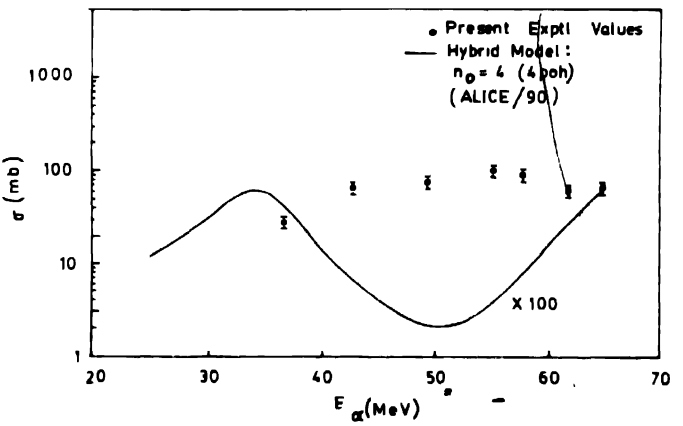


**Figure 2.** Excitation function of $^{169}Tm(\alpha,\alpha n)^{168}Tm$ reaction.

The only plausible explanation left out is the direct interaction effect. The bare, structure-less shape of the excitation function is itself a pointer to the direct reaction, namely, the inelastic scattering of the incident α-particle followed by a neutron evaporation. Similar observations were made by Blann and Lanzafame [16] while studying the α-induced reactions on gold, especially $^{197}Au(\alpha,\alpha n)$ reaction. In their work, they have studied the recoil ranges of the residual nuclei $^{196}Au$ and found that there is very little momentum transfer to recoiling nucleus (roughly about 10% of the momentum transfer for that a compound nucleus would have involved). On this basis, they have proposed the operation of considerable non-compound mechanism such as direct interaction and/or pre-equilibrium decay.

### *3.3. $^{169}Tm(\alpha,\alpha 2n)^{167}Tm$ reaction :*

Figure 3 shows the presently measured values of excitation function together with theoretical predictions of hybrid model for the initial exciton number $n_0 = 4(4p0h)$. There are isobaric precursor contribution in the study of this reaction with the result that the experimentally measured cross section at any energy is the sum of the cross-sections for the three reactions, $(\alpha,6n)$, $(\alpha,p5n)$ and $(\alpha,\alpha 2n)$ on the target $^{169}Tm$.

The weird shape of the excitation function with sudden exponential rise in the measured cross sections beyond 55 MeV, is clearly seen to be the result of isobaric contribution from the two interfering reactions (α, 6n) + (α, p5n). However, below 55 MeV

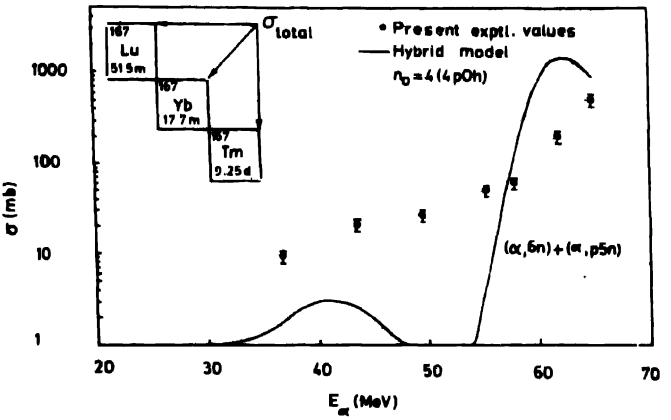


**Figure 3.** Excitation function of $^{169}Tm(\alpha,\alpha 2n)^{167}Tm$ reaction.

which is the effective threshold for (α,6n) and (α,p5n) reactions, the theoretical cross sections are that due to $^{169}Tm(\alpha,\alpha 2n)$ reaction. It can be seen that hybrid model predictions are lower by a factor of ten or even more. Furthermore, it is also interesting to see that the exponentially rising part of the excitation function is adequately accounted for, by the hybrid model predictions. This is because, this part of excitation function comprises mainly of the cross section of (α,6n) and (α,p5n) reactions of which later is approximately an order of magnitude smaller than the former.

### *3.4. $^{169}Tm(\alpha,\alpha 4n)\,^{165}Tm$ reaction :*

Figure 4 shows the excitation function for this reaction together with theoretical prediction of hybrid model using $n_0 = 4(4p0h)$ configuration. For this reaction also, although in principle the isobaric precursor contributions are possible as shown in the inset of the figure, in practice these contributions are very small. This is why there is no abrupt rise observed in the experimental measurements in this case as found in the previous ones. The shape of the excitation functions is reminiscent of the direct reaction model.

## 4. Conclusions

In the present work, alpha induced reactions on the target element thulium are investigated upto 60 MeV. Excitation functions for the reactions $^{169}Tm(\alpha,p3n)$, $^{169}Tm(\alpha,\alpha xn)$ ; $x = 1, 2, 4$ are studied in which isobaric contributions are observed. The observed isobaric contributions in the case of $^{169}Tm(\alpha,p3n)$ reaction is adequately accounted for, by using the formula developed from first principles. The corrected experimental cross sections for both

these reactions show a fair agreement with hybrid model predictions with $n_0 = 4$ $(4p0h)$ configuration. In the case of $(\alpha,\alpha xn)$ type of reactions, the model predictions are underestimated by more than an order of magnitude. A possible reason for the large experimental values may be the direct inelastic scattering of incident alpha followed by a neutron evaporation in this type of reactions.

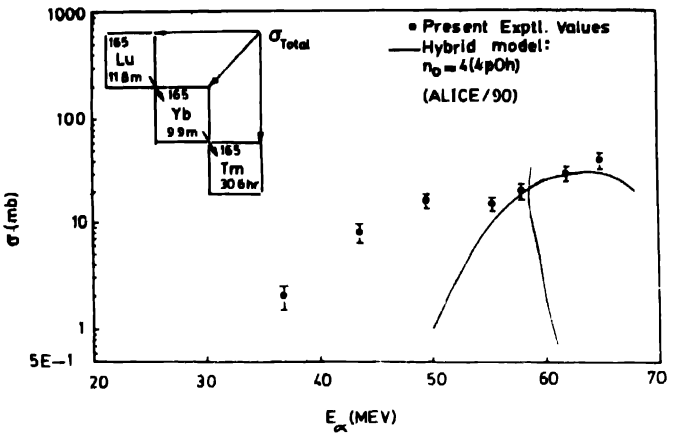


**Figure 4.** Excitation function of $^{169}Tm(\alpha,\alpha 4n)^{165}Tm$ reaction.

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## Appendix

Let $T$ be a target containing $N_T$ nuclei per unit area and $\phi$ be the flux of a particles incident on it for a period of time $t_i$. During this period of irradiation, two isobaric nuclei $B$ and $C$ are produced through $(\alpha,xn)$ and $(\alpha,px'n)$ reactions with cross sections $\sigma_1$ and $\sigma_2$ respectively. Also the isobaric nuclei are radioactive with disintegeration constants $\lambda_1$ and $\lambda_2$ respectively forming a decay chain,

$$\begin{array}{ccccc} T & \rightarrow & B & \rightarrow & C \\ \lambda_T & & \lambda_1 & & \lambda_2 \end{array}$$

with the notation that $T$ stands for target nucleus which is stable ($\lambda_T \cong 0$), $B$ and $C$ are mother and daughter nuclei respectively.

After a "Period of Irradiation" $t_i$ the products are allowed to decay for a waiting time "$t_w$" and then the activity of the daughter nucleus $C$ is measured after the time, "$t_w + \Delta$" starting from "$t_w$" then,

*Stage 0 :* Upto the end of irradiation. According to Batemann's [17] equations.

$$B_{t_i,o} = \frac{N_T\,\phi\,\sigma_1}{\lambda_1}\left(1-e^{-\lambda_1 t_i}\right) \qquad \text{(I)}$$

and

$$C_{t_i,o} = \frac{N_T\phi\sigma_1}{\lambda_2}\left\{1 + \frac{\lambda_2}{\lambda_1 - \lambda_2}\,e^{-\lambda_1 t_i} - \frac{\lambda_1}{\lambda_1 - \lambda_2}\,e^{-\lambda_2 t_i}\right\} \qquad \text{(II)}$$

**Stage 1 :** After the end of irradiation, at any time "$t$",

$$B(t) = B_{t_i,o}\,e^{-\lambda_1 t} \qquad \text{(III)}$$

with the condition that at $t = 0$, $B = B_{t_i,o}$. Further, the rate of decay of $C$ is given as

$$\frac{dc}{dt} = \lambda_1 B - \lambda_2 C$$

or

$$\frac{dc}{dt} + \lambda_2 C = \lambda_1 B \qquad \text{(IV)}$$

substituting the value of $B$ from equation (III) and multiplying equation (IV) by $e^{\lambda_2 t}$, we have

$$e^{\lambda_2 t}\frac{dc}{dt} + \lambda_2 C e^{\lambda_2 t} = \lambda_1 B_{t,o} e^{-\lambda_1 t} e^{\lambda_2 t}$$

or $$\frac{d}{dt}\left(C e^{\lambda_2 t}\right) = \lambda_1 B_{t,o} e^{(\lambda_2 - \lambda_1)t}$$

Integrating the above expression, we get

$$C e^{\lambda_2 t} = \frac{\lambda_1 B_{t,o}}{\lambda_2 - \lambda_1} e^{(\lambda_2 - \lambda_1)t} + K \qquad \text{(V)}$$

where $K$ is a constant.

At $t = 0$, $C = C_{t,o}$

then, $$C_{t,o} = \frac{\lambda_1 B_{t,o}}{\lambda_2 - \lambda_1} + K$$

or $$K = C_{t,o} - \frac{\lambda_1 B_{t,o}}{\lambda_2 - \lambda_1} \qquad \text{(VI)}$$

substituting the value of $K$, in equation (V),

$$C e^{\lambda_2 t} = \frac{\lambda_1 B_{t,o}}{\lambda_2 - \lambda_1} e^{(\lambda_2 - \lambda_1)t} + C_{t,o} - \frac{\lambda_1 B_{t,o}}{\lambda_2 - \lambda_1}$$

or $$\lambda_2 C(t) = \frac{\lambda_1 \lambda_2 B_{t,o}}{\lambda_2 - \lambda_1} e^{-\lambda_1 t} + \lambda_2 C_{t,o} e^{-\lambda_2 t} - \frac{\lambda_2 \lambda_1 B_{t,o}}{\lambda_2 - \lambda_1} - e^{-\lambda_2 t}$$

substituting the values of $B_{t,o}$ and $C_{t,o}$ from equations (I) and (II) ;

$$\lambda_2 C = \frac{N_T \phi \sigma_1 \lambda_2}{\lambda_2 - \lambda_1}\left[\left(1 - e^{-\lambda_1 t_i}\right) e^{-\lambda_1 t} - \frac{\lambda_2 - \lambda_1}{\lambda_2}\left(1 + \frac{\lambda_2}{(\lambda_1 - \lambda_2)}\right.\right.$$

$$\left.\left. e^{-\lambda_1 t_i} - \frac{\lambda_1}{(\lambda_1 - \lambda_2)} e^{-\lambda_2 t_i}\right) e^{-\lambda_2 t} - \left(1 - e^{-\lambda_1 t_i}\right) e^{-\lambda_2 t}\right]$$

$$\lambda_2 C_1 = \frac{N_T \phi \sigma_1 \lambda_2}{\lambda_2 - \lambda_1}\left[e^{\lambda_1 t}\left(1 - e^{-\lambda_1 t_i}\right) - \frac{\lambda_2}{\lambda_1} e^{-\lambda_2 t}\left(1 - e^{-\lambda_2 t_i}\right)\right] \qquad \text{(VII)}$$

and $$\lambda_2 C_2 = N_T \phi \sigma_2 \left(1 - e^{-\lambda_2 t_i}\right) e^{-\lambda_2 t} \qquad \text{(VIII)}$$

The reaction product $C$ can be formed in two ways, (i) by the decay of its isobar $B$ and (ii) directly in the reaction.

These two modes of formation of $C$ are given by equations (VII) and (VIII). Therefore, for total activity of $C$,

$$\lambda_2 C = \lambda_2 C_1 + \lambda_2 C_2$$

from equations (VII) and (VIII)

$$\lambda_2 C = N_T \phi \left[ \frac{\sigma_1 \lambda_2}{(\lambda_2 - \lambda_1)} \left\{ e^{-\lambda_1 t'} (1 - e^{-\lambda_1 t'_1}) - \frac{\lambda_1}{\lambda_2} e^{-\lambda_2 t'} \left(1 - e^{-\lambda_2 t'_1}\right) \right\} \right.$$

$$\left. + \sigma_2 (1 - e^{-\lambda_2 t'_1}) e^{-\lambda_2 t'} \right]$$

Integrating between $t_w$ and $t_w + \Delta$ and simplifying we get,

$$\frac{A_\gamma}{P_\gamma \theta_\gamma N_T \phi} = \left[ \frac{\sigma_1 \lambda_2}{\lambda_2 - \lambda_1} \left(1 - e^{\lambda_1 t'_1}\right) \left\{ \frac{e^{-\lambda_1 t'_w} - e^{-\lambda_1 (t_w + \Delta)}}{\lambda_1} - \frac{e^{-\lambda_2 t'_w} - e^{-\lambda_2 (t_w + \Delta)}}{\lambda_2} \right\} \right.$$

$$+ \frac{\sigma_1 + \sigma_2}{\lambda_2} \left(1 - e^{-\lambda_2 t'_1}\right) \left(e^{-\lambda_2 t'_w} - e^{-\lambda_2 (t_w + \Delta)}\right) + \frac{\sigma_1}{\lambda_2 - \lambda_1}$$

$$\left. \left(e^{-\lambda_1 t'_1} - e^{-\lambda_2 t'_1}\right) \left(e^{-\lambda_2 t'_w} - e^{-\lambda_2 (t_w + \Delta)}\right) \right] \qquad \text{(IX)}$$

The quantities on LHS of equation (IX) can be written as

$$\frac{A_t A_\gamma}{\phi \theta_\gamma P_\gamma P_t w_t N_{av}}$$

$A_\gamma$ – photo peak area of characteristic $\gamma$ ray with abundance $\theta_\gamma$ and the efficency of the detector being $P_\gamma$.

$P_t$, $A_t$, $w_t$ and $N_{av}$ are isotopic abundance, mass number, weight of the foil per unit area and Avagadro Number respectively.